Appendix A

Improving Thermal Conductivity of Plastics

Introduction

The purpose of this invention is to improve the thermal conductivity of plastics through solvent blending techniques. Improved thermal conductivity depends on both the matrix materials, and the fillers used. Currently used fillers include metals such as aluminum, copper, nickel, boron nitride, beryllium oxide, carbon, graphite, etc... These fillers are used in the shape of powder, fiber, and flake. The degree of improvement depends on such factors as the thermal conductivity of the filler and, in the case of fibers and flakes, in maintaining maximum length and orientation.

A unique new grade of carbon fibers was recently introduced to the market by Amoco Corporation, ThermalGraph DKA-X and ThermalGraph DKD-X. These fibers have unusually high thermal conductivity. Because these new fibers have unusually high bulk factor and are unusually sensitive to fiber break-down, they present special problems in blending and molding.

This invention describes methods to incorporate these materials in high filler content into plastics and also to maintain fiber length. The fibers are added to a solution of plastic with minimum of stirring to improve wettability, dispersability, and shape retention. This type of encapsulation also improves physical properties.

The method is also used with other fillers such as boron nitride powder which is extremely bulky.

The method is also useful in making concentrates of fibers and other fillers which can be easily let down to tailor-make materials of a given thermal conductivity and physical property.

For both ThermalGraph and boron nitride, we have made concentrates containing 90% by weight. These concentrates are useful for plastics which are not soluble in any solvent.

The method of blending involves dissolving Ultem 1010 in methylene chloride and then adding the thermalgraph material and/or boron nitride. The mix is stirred slowly until the fibers are completely wetted out. Stirring is continued while the methylene chloride evaporates. Methylene chloride evaporates rapidly. After the mix has thickened, it is poured onto a flat pan and placed in an oven at 300°F. Drying occurs quickly. Granulate the resin and injection mold.

Another solvent used in N methyl pyrrolidone. This solvent, as with methylene chloride, has excellent wetting characteristics. This solvent can be removed by pouring into water. The solvent is very water soluable.

An average batch consisted of 100 grams of Ultem 1010; 400 grams of filler; and 900 grams of methylene chloride, yielding 500 grams of blended material.

Table 1 lists the compounds tested and the method of blending.

Description of Raw Materials Used in This Invention

Ultem 1010: High flow, low molecular weight polytheremide manufactured by General Electric Corporation.

Victrex Peek Grade 150: High flow, low molecular weight polyetheretherimide manufactured by the Victrex Corporation.

PPS: Polyphenylene Sulfide; product #020584, natural; manufactured by Ticona Corporation

ThermalGraph DKA-X: High thermal conductivity fibers manufactures by the Amoco Corporation. Thermal conductivity in the fibers disection equals 800-1,100 W/M/°K, filament diameter equals 10 microns; average length equals 200 microns.

ThermalGraph DKD-X: Same as DKA-X above except that the thermal conductivity equals 300-700 W/M/°F.

Boron Nitride: Carbotherm CTFS; boron nitride powder manufactured by the Carborundum Corporation; mean particle size is 5-10 microns.

Aluminum Flake: Transmit K102 manufactured by the Transmit Corporation; 1mm.X 1.3mX30 microns particle size;0.32 grams/cc bulk density.

Aluminum Pigment Concentrate: Solvent 110-20-E manufactured by Silberline Manufacturing Corporation Incorporated. 80% aluminum pigments and 20% resin; particle size of the pigments is 23 microns.

Methylene Chloride: Technical grade methylene chloride manufactured by the Dow Chemical Corporation.

N Methyl Pyrrolidone: Technical grade N methyl pyrrolidone manufactured by BASF Corp.

Description of Molding Techniques

Molding of materials has to be done with great care with the ThermalGraph materials in order not to reduce particles length. Longer fibers make better thermal conductivity.

When molding with a screw injection molding machine care must be taken to reduce shear as much as possible by use of a large nozzle , a zero compression screw if possible, large runners and large gates.

Best results are obtained with an old fashion ram injection molding machine which has the minimum amount of shear.

It is critically important in a production run to check the density of the parts. Parts can look satisfactory but have a low density resulting in as much as fifty -percent reduction in the thermal conductivity.

Description Of Test Method

The following equipment and specimens are used:

1) A heat sink- we use a Digis Block manufactured by Laboratory Devises Inc.

2) A temperature controller- We use a Modbus controlled temperature controller manufactured by the presys. Corporation with a "T" type probe.

3) A lap top computer with Excel and a program in Excel called "the Wedge".

4) Plastic specimens measuring 2¼ × ½ × ¼. The specimens are grounded flat on both sides.

5) A thick thermally conductive paste containing boron nitride and white oil

The heat sinks is set with a ΔT of 40°C above room temperature. The bottom of the test specimen is covered with a conductive paste to improve thermal contact. A file is prepared on Excel; conditions set on "the wedge" program; the wedge turned on; and the specimen and the thermocouple placed on the heat sinks. Temperature measurements are placed into column one of the spreadsheet at the rate of two reading per second. The thermocouple and the sample are withdrawn after approximately 30 seconds and the Wedge program halted. The sample data is graphed. The first 10 to 15 readings form a constant slope. A linear equation is automatically plotted and the slope recorded. This yields a rate of increase °/sec, calculated at 2 × the reading (two reading are taken per second.)

This rate of temperature increase, °C/sec, This correlates very well with thermal conductivity. It is a much faster way of rating the thermal conductivity of plastics up to approximately 7W/M/°K beyond that the test is not accurate.

Discussion of Results

The most common way of making high performance plastic compound is to use a twin screw extruder with a pelletizer. The new materials can be either pre-blended or introduced at separate feed zones in the extruder. The high sheer stresses in the twin screw extruder, which are good for mixing, break down fiber length of the thermograph fibers thereby reducing thermal conductivity. The twin screw extruder can not make moldable compounds above 50% concentration. The wettability and dispersability of the fillers in the melt stage is much less than in the solution. Wettability and dispersability depend on the ability of the plastic to encapsulate and separate individual particles of filler. The end result of good wettability and despersability is that the filler is much more effectively used and the thermal conductivity thereby improved. Methylene chloride and N methylene pyrrolidone are solvents which very effectively cause these improvements. Table One demonstrates the large improvements in thermal conductivity by solvent blending.

TABLE ONE: Comparison of Solvent Blended vs. Dry Blended and Extrusion Blended Compounds for Thermal Conductivity Using Mack Plastic Tests.

Sample Designation	Method of Blending	Test Results	
		Microtensile Sample	2½"×½"×¼" Sample
1010/DKD (50/50)	Solvent		1.326
1010/DKD (40/60)	Solvent		1.792
1010/DKD (30/70)	Solvent	3.075	3.516
PPS/DKD (50/50)	Dry blend		.926
PPS/DKD (40/60)	Dry blend		1.416
PPS/DKD (30/70)	Dry blend	2.525	2.211
PEEK/DKD (30/70)	Extrusion	2.501	
1010/DKD/BN (50/25/25)	Solvent		1.140
PEEK/DKD/BN (50/25/25)	Extrusion		.684
1010/BN (50/50)	Solvent	2.137	1.072
PEEK/BN (50/50)	Extrusion	1.492	.759
1010/K102 (50/50)	Solvent		1.570
PPS/K102 (50/50)	Dry blend		1.229
PPS/K102 (50/50)	Extrusion		.777
1010/Graphite (AGM 3243) (50/50)	Solvent	2.897	
PPS/Graphite (AGM 4390) (50/50)	Dry blend	2.497	